Xe(L) x-ray emission from laser-cluster interaction

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Abstract

We have measured absolute L x-ray emission yields from $(Xe)_n$ clusters (with n in the range $10^5 – 10^7$ atoms/cluster) irradiated by 60 femtoseconds 800 nm IR and 160 femtoseconds 400 nm UV laser pulses of $10^{15} – 10^{17}$ W cm^{-2} peak intensity. Measurements have been performed as a function of cluster size (backing pressure) and laser peak intensity. Identification of spectroscopic features as well as x-ray emission yield variation with laser wavelength and intensity are in strong contradiction with results and interpretation from previous studies.

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1. Introduction

KeV x-ray emission can be produced from rare-gas clusters by irradiation with intense (I ~ 10^{15}–10^{18} W cm^{-2}) femtosecond (t ~100 fs) laser pulses : for instance, Ar(K), Kr(L) and Xe(L) have been observed [1-3]. In the case of Xe clusters, previous studies [4] have shown a strong dependence as a function of laser wavelength. However, such a dependence was not observed in the case of Ar clusters, where an extensive study has been performed [3]. As the mechanisms for the production of inner shell ionisation remain controversial, quantitative measurements under well-controlled conditions and for a large range of variation of the various parameters governing the interaction are still needed. We have quantitavely measured L x-ray emission from Xe clusters of various sizes in the range 10^5-10^7 atoms/cluster, for infrared (800 nm) and ultraviolet (400 nm) pulses of 60 fs duration (160fs for UV), and as a function of laser peak intensity in the range 10^{15} – 10^{17} W cm^{-2}. Our results do not agree with those of previous studies [4].

2. Experiment and results

The experimental set-up, including the apparatus used for cluster generation and the X-ray spectrometers, has been described in detail in previous publications [3,5]. Briefly, Xe clusters are generated within a pulsed adiabatic expansion of the well documented Hagena type [6]. This device leads to a mean cluster size $\tilde{N}_c \approx 2.3 \times 10^4 P_0^{1.95}$ where P_0 is the backing pressure (bar). The intense laser field is generated with a Ti:sapphire laser system delivering pulses of minimum duration of 50fs at 800 nm with a repetition rate of 20Hz. The beam diameter is approximately 45mm and the maximum pulse energy available in the interaction zone is 90mJ. The laser light is focused with a f = 480 mm lens leading to maximum peak intensities ~ 10^{17}Wcm^{-2}.

The X-rays are analyzed using two Si(Li) detectors and a high-resolution high-transmission Bragg-crystal spectrometer equipped with a flat mosaic graphite crystal (HOPG) and a position sensitive detector working in the photon counting mode. All three detectors have well known transmission and efficiency. In some cases, thin absorbing Fe windows have been used to reduce counting rates. Their transmission has been carefully measured, and spectra shown here have been corrected for window absorption when necessary.

The Si(Li) detectors allow observation of the complete x-ray spectra in the region of Xe L transitions. Fig. 1. shows a sample spectrum recorded for a 9. 10^{15} W cm^{-2} peak intensity at 400 nm and a 10.7 bar backing pressure. We always observe only two components, here centred at 4277 and 4600 eV (2.899 and 2.695 Å).

The spectral features are better resolved with the crystal spectrometer. Fig. 2. shows two spectra recorded for 3.4 10^{16} W cm^{-2} IR laser pulses at two different backing pressure (3.1 and 10.7 bar). We observe two broad components, with quite different shapes for the two backing pressure: a strong, non-linear, cluster size (backing pressure) dependence is observed, both on spectral shape and absolute x-ray emission yield. For higher backing pressure (larger clusters) we observe an enhancement of high energy (high charge states) components. The backing pressure dependence of the total L x-ray yield has been measured in the range 1-20 bar and found to follow a P^{α} law with $\alpha \geq 5/2$, in contradiction with the assumed linear dependence used by Kondo *et al.* [4]. Fig. 3. similarly shows two spectra recorded for 9. 10^{15} W cm^{-2} UV laser pulses at two different backing pressure (5.5 and 10.7 bar). We also observe here two broad components, but with similar shapes at the two pressure, both however different from the case of IR pulses. On the other hand, our spectra look similar to those of Kondo *et al.* [4].

We have also measured the x-ray yield dependence on the laser pulse energy, W_{las}. Our results are well accounted by a $W_{las}^{3/2}$ power law, with an intensity threshold for the appearance of L x-rays of ~ 2. 10^{15} W cm^{-2}, a result similar to our previous findings in the case of Kr and Ar clusters [3]. Once again, the linear dependence assumed by Kondo *et al.* [4] appears to be erroneous.

3. Discussion

We have perform Dirac-Fock (MCDF) calculations to interpret our spectra. Kondo *et al.* [4] have assign the two group of lines to 3d $\rightarrow$2p transitions in multicharged ($25 \leq q \leq 35$) Xe ions with one or two 2p vacancies. The production of such multiple inner-shell vacancies has been interpreted [7,8] as due to a “coherent, ordered many-electron motion” acting as a single heavy, highly charged, particle. From our calculations, we find however that the energy difference between transitions involving one or two 2p vacancies amounts to ~ 120 eV (0.08 Å) on the average, a value much too small to account for the observed energy difference between the two groups of transitions. Moreover, we find that the relativistic splitting between 3d5/2-2p3/2 and 3d3/2-2p1/2 transitions in multicharged (24+-32+) Xe ions is ~ 320 eV (0.20 Å), in good agreement with the observed splitting. We then conclude that there is in fact no evidence for the production of double 2p vacancies in the spectra. The observed structure correspond well to 3d $\rightarrow$2p transitions in multicharged ($24 \leq q \leq 32$) Xe ions with one 2p vacancy and various defect configurations and couplings in M shell; 3s $\rightarrow$2p transitions are not observed, but they may be embodied in the bremsstrahlung background on the low energy side (see fig. 1.). More surprisingly, there is little evidence of 3p $\rightarrow$2s transitions, which should appear on the high energy side (fig. 1.). Fluorescence and Auger yield, as well as Coster-Kronig in L subshells, have however to be calculated before one can conclude on possible selection rules.

We observe a strong spectral shape wavelength dependence, with higher charge states for the case of 400 nm radiation. However, contrarily to previous studies [4] where a λ^6 dependence was reported, only a weak wavelength dependence of the absolute emission yield is found : for identical laser pulse energy and backing pressure, the UV yield is only twice as large than with IR radiation. One possible reason for our disagreement with the conclusions of the above mentioned studies may be found in our observation of a strong non-linear dependence on both the cluster size and the laser intensity.

4. Conclusion

Absolute yields have been measured for Xe clusters irradiated by IR and UV laser pulses. Dependencies on backing pressure (cluster size), laser energy and wavelength have been studied. Our results contradict previous results [4,7,8] both on line assignments and wavelength dependency. An extensive theoretical work is needed before interpretation of the observed spectral shapes, intensity threshold and possible selection rule on vacancy production can be made.

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Figure captions

Figure 1.

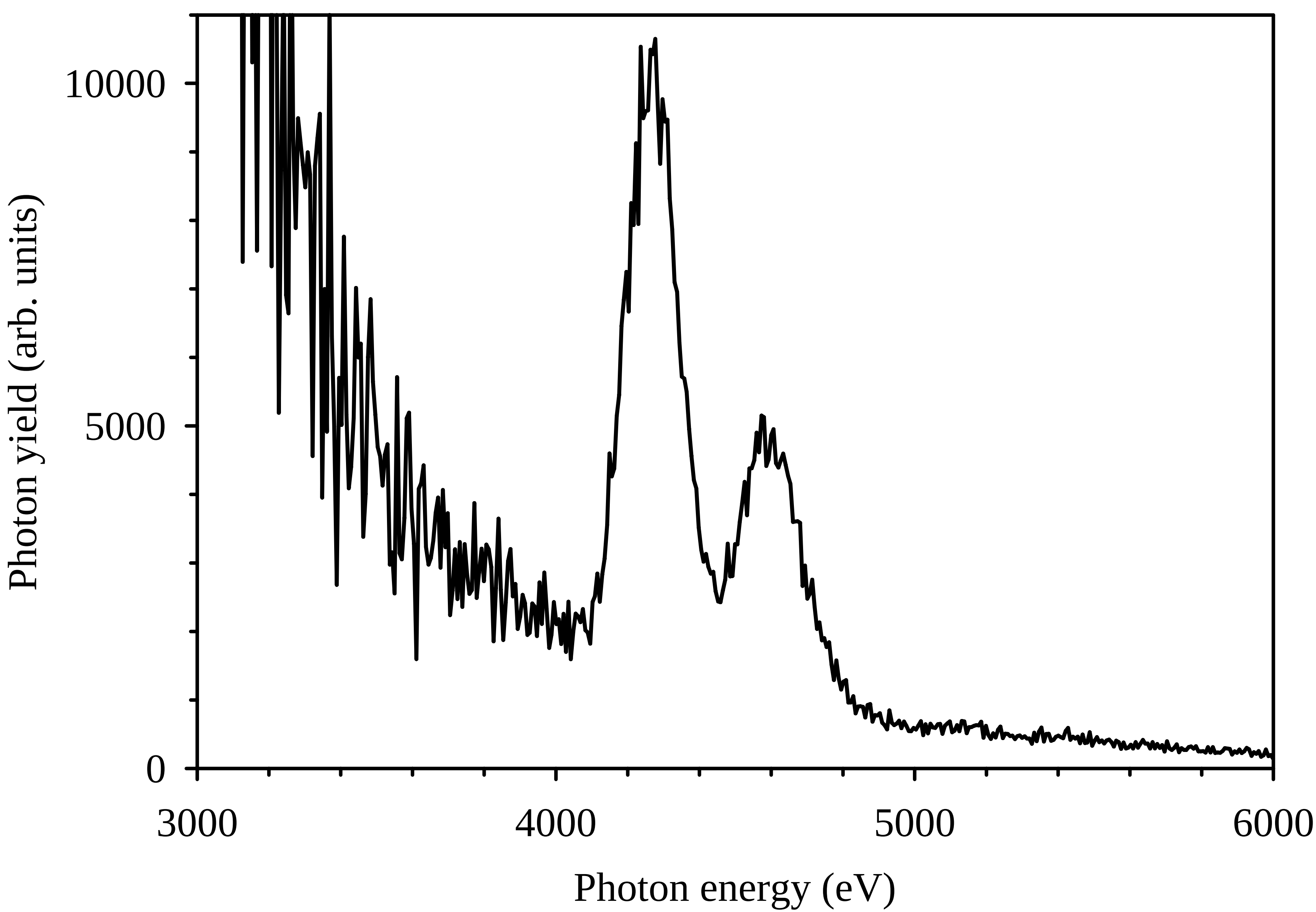
Si(Li) x-ray spectrum recorded for a 9. 10^{15} W cm^{-2} peak intensity at 400 nm and a 10.7 bar Xe backing pressure. Spectrum has been corrected for window transmission and detector efficiency.

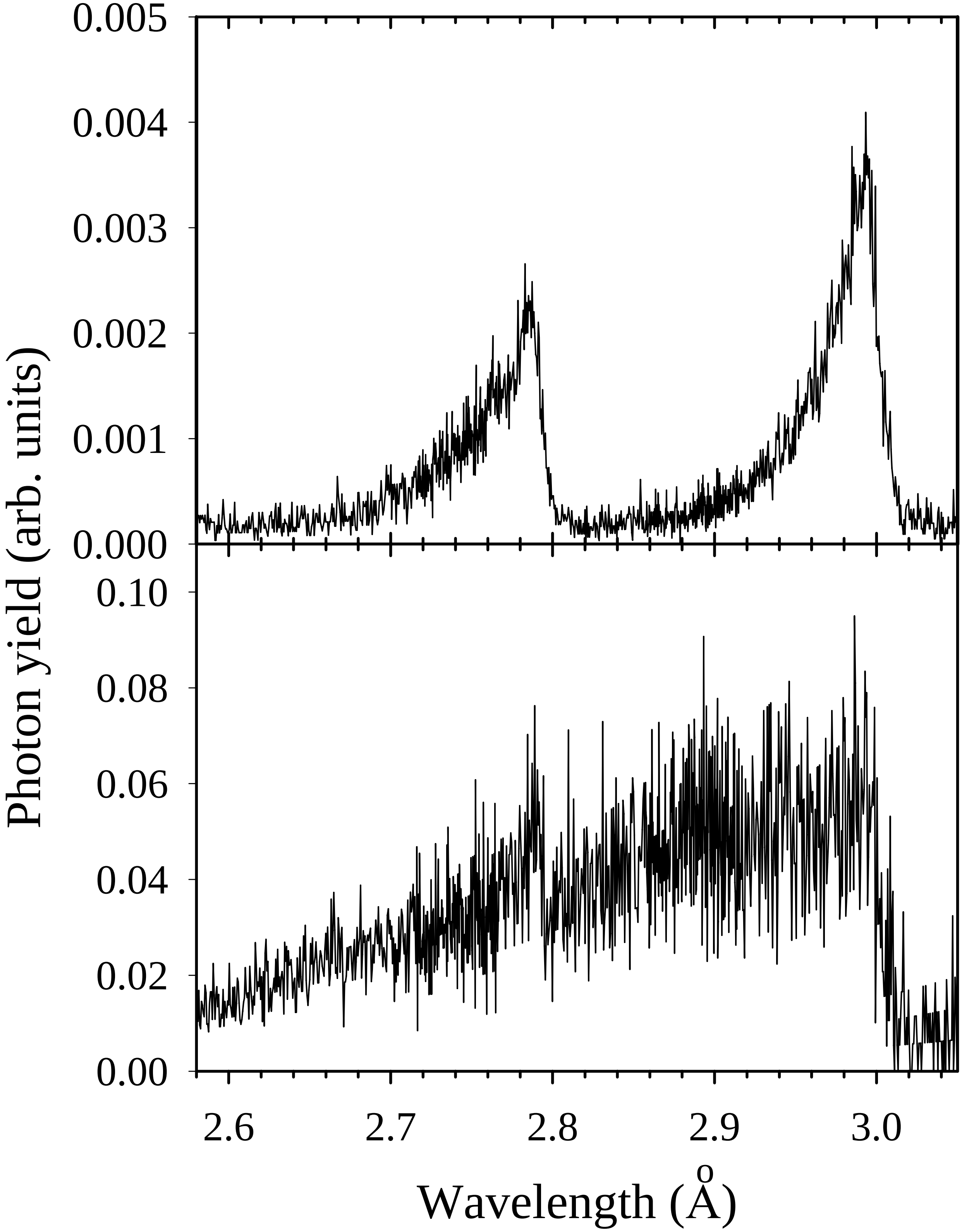
Figure 2.

Cristal spectrometer x-ray spectra recorded for 3.4 10^{16} W cm^{-2} IR (800 nm), 60fs, laser pulses at two different Xe backing pressure : a) 3.1 bar and b) 10.7 bar. Spectra have been corrected for window transmission and detector efficiency.

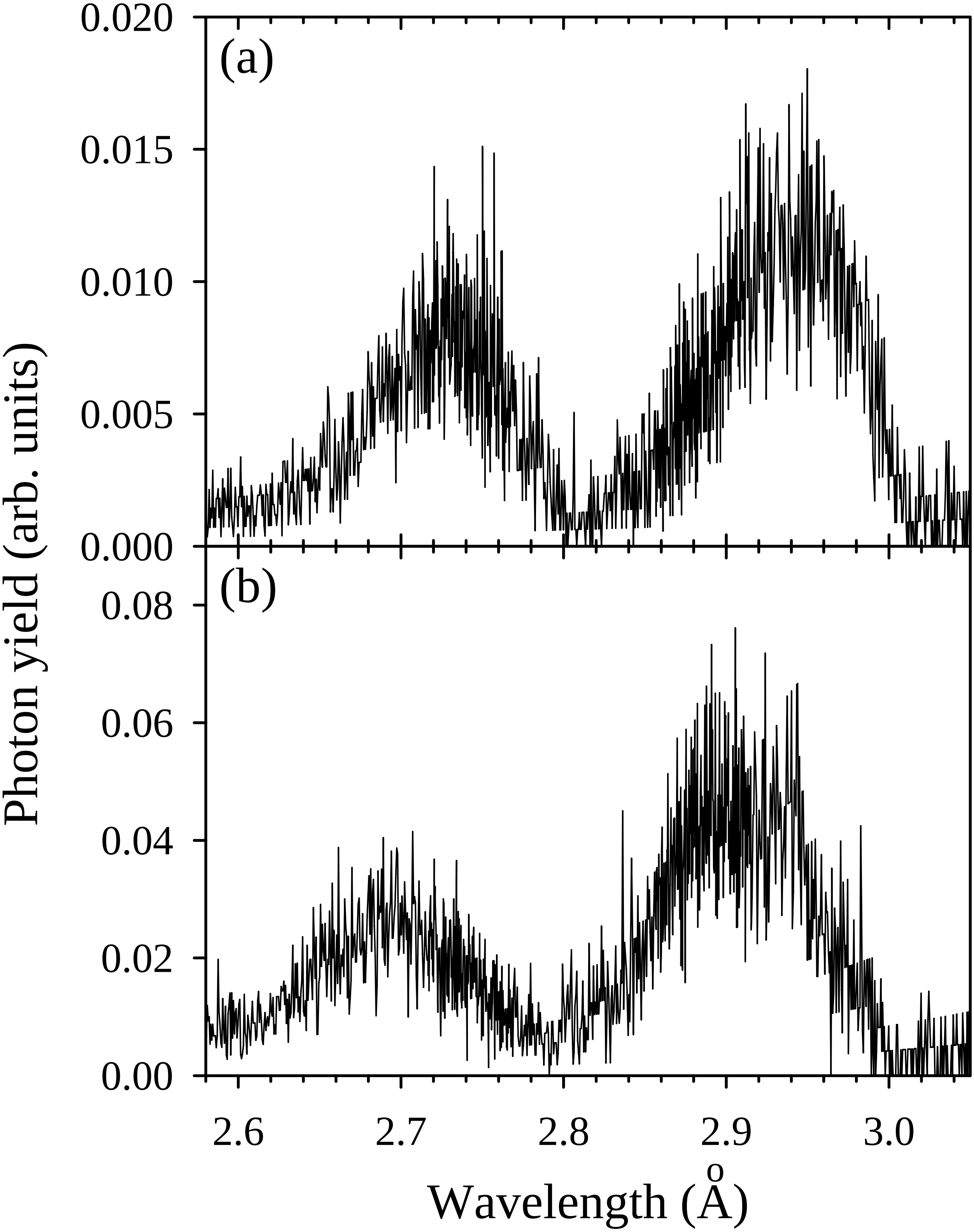
Figure 3.

Cristal spectrometer x-ray spectra recorded for 9. 10^{15} W cm^{-2} UV (400 nm), 160 fs, laser pulses at two different Xe backing pressure : a) 5.5 bar and b) 10.7 bar. Spectra have been corrected for window transmission and detector efficiency.


10000
5000
0
3000
4000
5000
6000
Photon yield (arb. units)
Photon energy (eV)



0.005
0.004
0.003
0.002
0.001
0.000
0.10
0.08
0.06
0.04
0.02
0.00
Photon yield (arb. units)
2.6
2.7
2.8
2.9
3.0
Wavelength (Å)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0.010
0.005
0.000
(a)
(b)
0.08
0.06
0.04
0.02
0.00
2.6
2.7
2.8
2.9
3.0
Photon yield (arb. units)
Wavelength (Å)